The optimisation of fingermark enhancement by VMD and Lumicyano™ on thermal paper

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Highlights

- Application of Lumicyano™ and of Au/Zn and Ag/Zn VMD on thermal paper.
- Different thermal papers required different amounts of gold and silver.
- Optimised Au/Zn and Ag/Zn varied in effectiveness on different thermal papers.
- Ag/Zn outperforms Au/Zn on fingermarks early in depletion series.
- Generally lower amounts of gold needed for enhancement than currently recommended.

Abstract

The enhancement of fingermarks on thermal paper can be challenging due to background staining caused by polar solvents used in fingermark enhancement techniques such as ninhydrin. This study explored a commercial one-step superglue fuming process, Lumicyano™, and Vacuum Metal Deposition (VMD) to develop fingermarks on this substrate and overcome this issue. Different sequential treatments involving Lumicyano™ and a combination of VMD methods were investigated with varying degrees of success with some sequences being highly sensitive. The VMD processes, however, were observed to generally be more effective at enhancing marks, whereas Lumicyano™ provided little or no benefit on this paper type. The results indicate that Lumicyano™ is only beneficial as a pre-treatment when the entire sequence of gold/zinc and silver/zinc is taken to completion. The gold/zinc and silver/zinc VMD processes were optimised on five different thermal papers, and the optimised techniques were then directly compared to determine which was more successful on each thermal paper type as a single treatment.

Keywords: Latent fingerprint enhancement, thermal paper, Vacuum Metal Deposition, solvent free, one step superglue fuming, environmentally friendly techniques

1. Introduction

Thermal papers were developed in 1968 by the National Cash Register Company [1] and consist of a number of layers, including a thermally sensitive active layer. This thermally active layer includes a leuco dye which changes from the colourless to coloured form when an external stimulus such as heat is applied and gives the paper its desired properties. Commonly, these turn black when heat is applied, although the addition of un-substituted fluoran compounds during production can result in different colours being produced [2]. Due to the inexpensive and rapid nature of printing, these papers are commonly used in everyday life as point of sales receipts [3]. However, common fingermark (or latent fingerprint) enhancement techniques which can be employed for paper result in a blackening of the substrate leading to a loss of contrast of the mark as well as other printed document evidence. Polar organic solvents in the development techniques cause premature oxidisation of the dye present in the paper which would normally have been oxidised by an acid present when subjected to thermal stimulus [4]. To prevent this blackening, the Fingermark Visualisation Manual (FVM) [5] recommends stripping the thermally active layer with acetone prior to enhancing fingermarks; however, this removes any printed evidence and may be detrimental to subsequent fingermark or forensic processing [5]. In addition to the blackening, the European regulation on the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), regulates the use of potentially harmful chemicals in the EU [6] including solvents. Due to these factors, alternative, environmentally friendly methods of enhancing fingermarks on this challenging substrate are required.

The structure, topography, chemistry, surface energy and porosity of paper varies with type, brand and origin [36]. It is shown by [7,8] that variation of topography, even within a single class of substrate, affects fingermark development. The porosity of a substrate is commonly taken into account when selecting the most appropriate enhancement technique to use and must be considered when investigating thermal paper as this substrate is both porous and semi-porous due to the layers used in manufacture.

The recommended processes for visualising fingermarks on thermal paper varies internationally. In the UK for example, current guidelines involve a pre-treatment with acetone to strip the thermal layer followed by the normal sequence of treatments for paper (indanedione then ninhydrin). This process is successful at enhancing marks; however, the stripping of the thermal layer removes text, causes any written evidence on the paper to be lost and if the article is not processed in acetone long enough to fully remove the thermosensitive layer, blackening can still occur [5,9]. Additionally, the carrier solvent HFE7100, frequently used in these processes is relatively expensive (£85.33 per litre as of 2017), has a high Global Warming Potential (GWP), and the shelf-life of the prepared solution is approximately only 1 week [4,10–13]. These problems also occur with the acid-free indanedione standard method employed by the Autralian Federal Police (AFP) for thermal paper. Alternatives have been developed which aim to avoid this blackening through different means. A solvent-free example of development via amino acid reagents [14] uses paper impregnated with indanedione to enhance marks on thermal paper. This process uses a dry contact approach at room temperature for 48 hours and reported results that were comparable or better than the standard AFP method. Additionally to avoid background blackening, vacuum vaporisation of ninhydrin can be used [15]. This technique requires no solvents or heat to be applied to the thermal paper.

Alternatively, research has investigated incorporating G3, a petroleum based mixture of amides and amines, into the indanedione development process as a post-treatment [12]. This can be very successful at reversing the blackening caused by polar solvents or heating of the thermal paper; however, the darkening can return over time [12]. Fitzi *et al*. [16] investigated a method using 1,4-diazabicyclo[2.2.2]octane (DABCO), which acts by a Lewis base modifying the leuco-dye structure to reverse darkening of the paper in the same way as G3. Whilst both solutions were effective at removing the blackening on most thermal papers, G3 achieved slightly better quality of marks in trials. However, the authors recommended the use of DABCO as it was less expensive, easier to prepare and had a lower toxicity than G3. In a similar way, to overcome darkening, polyvinylpyrrolidone (PVP) has been incorporated into the ninhydrin method and has been seen to be successful at enhancing marks by reducing the blackening typically caused by this process [17]. The drawback of this technique is that inks present on the thermal paper can dissolve due to the ratio of polar to non-polar solvents (1:9) however staining of the background was prevented and developed marks remained stable for six months.

In addition to the altered amino acid targeting methods, other techniques have been developed to overcome the blackening issue. The controlled application of heat to enhance latent marks has been explored [18–22] and whilst this removes the necessity to use polar or harmful chemicals, as reported by Bond *et al*. [22], the temperature required varied on thermal paper from different regions as did the effectiveness of the technique. Another method that uses the intrinsic properties of the thermally active layer is fuming with hydrochloric acid [23,24].

Whilst this method is inexpensive, and print development is rapid, the studies examining hydrochloric acid accept that the process is highly corrosive, precludes other enhancement methods and has inconsistent results.

There are numerous other techniques available for the enhancement of fingermarks, including cyanoacrylate fuming and Vacuum Metal Deposition (VMD) [5]. Lumicyano™, a commercial product based on cyanoacrylate fuming, is a one-step technique that incorporates a fluorescent dye into the cyanoacrylate-based liquid prior to heating [25]. It has been found to have an equal or greater sensitivity to mark enhancement than cyanoacrylate fuming, followed by BY40 [26,27]. The water component of the fingermark residue, being a weak base, initiates the growth of the cyanoacrylate polymer present in Lumicyano™ [28]. It is also suggested that the high relative humidity of 80% is sufficient for sodium chloride (NaCl) crystals present in the residue to take up water, which may initiate and promote polymerisation [29]. A recent study which compared Lumicyano™ to black magnetic powder [30] found that this Lumicyano™ can be effective on thermal paper.

The first recommended use of VMD was by Kent *et al*. [31]. This technique develops latent fingermarks by first depositing a metal (typically gold) onto the surface by thermal evaporation under vacuum followed by the evaporation of zinc. The gold deposits onto the surface of the substrate and begins to form islands of gold. These have growth rates and morphology which depends upon various factors such as the topography, surface energy and chemical species present. This discontinuous layer of deposited metal is only a few nanometres thick. These gold particles, deposited on the fatty acid residue or ridges, become buried to a depth which makes them unavailable at the surface which precludes zinc deposition. Therefore, zinc is deposited on the substrate, or the valleys of the print, and not on the ridges which leads to a tendency to observe ‘negative’ development of latent marks [32]. Alternatively, the development observed could be attributed to the zinc growing at different rates on areas of different nucleation site size and morphology [9]. Alternative VMD processes which are also employed include silver and silver/zinc [33] however the FVM [5] states that silver VMD should only be used after gold/zinc in treating items sequentially. The VMD technique has the advantage over many of the aforementioned techniques in that it requires no solvents, which ensures there is no loss of evidence from the thermal paper due to blackening.

This investigation examined the potential of gold/zinc (Au/Zn) VMD, silver/zinc (Ag/Zn) VMD, silver (Ag) VMD and Lumicyano™ in sequence to determine the most effective combination of treatments whilst assessing the sensitivity of each technique. Subsequently, the VMD methods deemed to be most effective were optimised on different thermal papers whilst the reproducibility of the Ag/Zn process was increased. Finally, a comparison study determined which of the optimised VMD techniques enhanced fingermarks most effectively on these thermal paper types.

2. Method

2.1 Study 1: Sequential Treatment

2.1.1 Thermal Paper

The thermal paper used in this study had not been subjected to thermal printing and was produced by Eposgear™. All samples were prepared according to Figure 1 to allow for a 10 mark depletion series of natural, ungroomed fingermarks (i.e. the donors performed no action to actively increase the residue present on their fingertips). These marks were deposited on the thermally active side of the thermal paper by each of six donors who were instructed not to wash their hands for at least 30 minutes prior to depositing marks. This is similar to previous studies [34,35] and is based on the International Fingerprint Research Group (IFRG) guidelines [36]. To achieve a split depletion series, each donor was requested to touch the paper with a firm and consistent pressure with their right forefinger. Then, without reloading the same finger, they touched the box beneath in the same manner (Figure 1), until a 10 mark depletion series was achieved. The donors' propensity for depositing good or poor fingermarks was not known prior to the experiment, and the donors themselves were a mix of males and females between the ages of 21-55.

The marks were aged for one week at room temperature and humidity, as this was a reasonable ageing condition consistent with those described by the IFRG [36]. Following the ageing period, the samples were cut down the dashed line and enhanced using a sequence of development techniques as detailed in Table 1.

2.1.2 Lumicyano™ processing

This single step cyanoacrylate fuming process was undertaken according to manufacturer's guidelines whereby an 8% w/w was used. To achieve this, 0.16g ± 0.01g of Lumicyano™ powder (Crime Scene Technology) was measured and dissolved in 2.0g ± 0.01g of Lumicyano™ solution in a new foil dish. An Air Science CA305 cyanoacrylate processing chamber was used to heat the Lumicyano™ solution at approximately 120°C for 60 minutes whilst at 80% humidity.

2.1.3 Vacuum Metal Deposition (VMD) processing

The VMD processing was undertaken using a VMD 360™ (West Technology) system. Gold wire (Alfa Aesar), 0.25mm in diameter and of 99.999% purity, was cut to 3mm as measured by eye against a ruler as recommended by the equipment manufacturer and FVM [5,33]. Silver wire (Alfa Aesar), of 99.9% purity and 0.5mm in diameter was cut to the manufacturer recommendation of 3-5mm. For processes that required zinc, shot of 1-5mm in size and 99.999% purity (Alfa Aesar) was used.

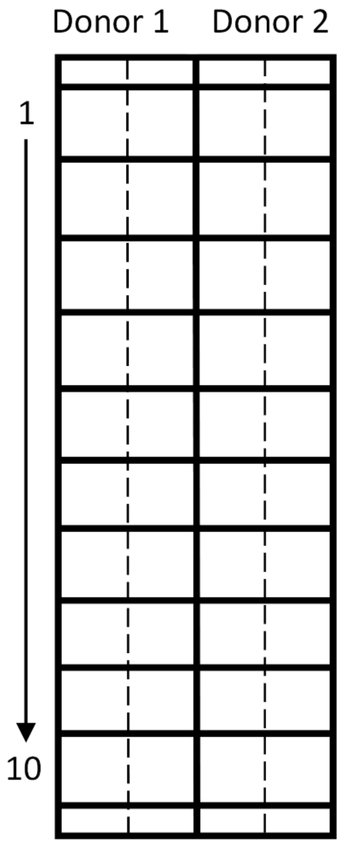

Figure 1: Diagram representing a single piece of thermal paper divided in two to enable donors to deposit side by side from depletion 1 to 10. Note the dashed line denoting where the paper was cut to obtain split marks.

Table 1: The four different sequences of treatments used in this study to identify the most effective position of Lumicyano™.

	Sequence 1	Sequence 2	Sequence 3	Sequence 4
Process 1	Au/Zn VMD	Lumicyano™	Au/Zn VMD	Lumicyano™
Process 2	Ag VMD	Au/Zn VMD	Ag/Zn VMD	Au/Zn VMD
Process 3	Lumicyano™	Ag VMD	Lumicyano™	Ag/Zn VMD

The chamber was evacuated to at least 2.2×10^{-4} mbar at which point the first metal (gold or silver) was slowly heated until evaporation was achieved. For the gold/zinc process, the entire piece of gold was evaporated, however, for the silver methods in this first study, heat was maintained until development, or a colour change was observed as described in the FVM [5]. Where zinc was part of the process, once the initial metal had been applied, the chamber pressure was automatically adjusted to $3 – 5 \times 10^{-4}$ mbar and zinc slowly evaporated from the second boat until development of the latent marks was observed. If a second process was required, then the items were removed, fingermarks graded and returned to the chamber for another process. The Ag/Zn technique was performed in the same way as the Au/Zn method without removing the sample from the chamber between the application of Ag and Zn. To ensure the successful deposition of the metals during the VMD procedure, a control of printer paper with a fresh mark from the investigator was developed at the same time as the samples. Development with Ag alone was carried out by thermally evaporating the Ag at 2.2×10^{-4} mbar until development of the latent marks was observed [5]. The VMD techniques were always applied in the order of gold/zinc before silver as outlined in Table 1, as recommended by the fingermark visualisation manual [5].

2.1.4 Visualisation and Grading of the Developed marks

The Lumicyano™ fluorescence was observed using a Quaser 2000/30 with blue/green fluorescence (band pass filter 468-526 nm at 1% cut-on and cut-off points respectively). This was viewed with an orange long pass 529 nm filter (1% cut-on point) and photographed using a Nikon D5100 digital SLR. White light, by varying the angle of illumination through manipulation of the sample by the observer, was used to improve the contrast of the VMD developed marks. The fingermarks were graded using the CAST system (Table 2) [36]. All grading of the half marks was carried out on the actual marks, rather than photographs, as soon after development as possible after each processing step by one examiner. Examples of the marks for each of the grades awarded can be seen in Figure 2.

Table 2: The grades awarded to the developed marks based on the CAST grading system with a definition and grade. The classifications, boxed in bold, are known as "identifiable fingermarks."

Classification	Definition
Grade 0	No mark has been developed
Grade 1	An empty mark has been developed
Grade 2	Some ridges have been developed with up to 1/3 of the mark having been developed
Grade 3	**Between 1/3 and 2/3 of the ridges of the mark have been developed**
Grade 4	**From 2/3 to a full mark has been developed**

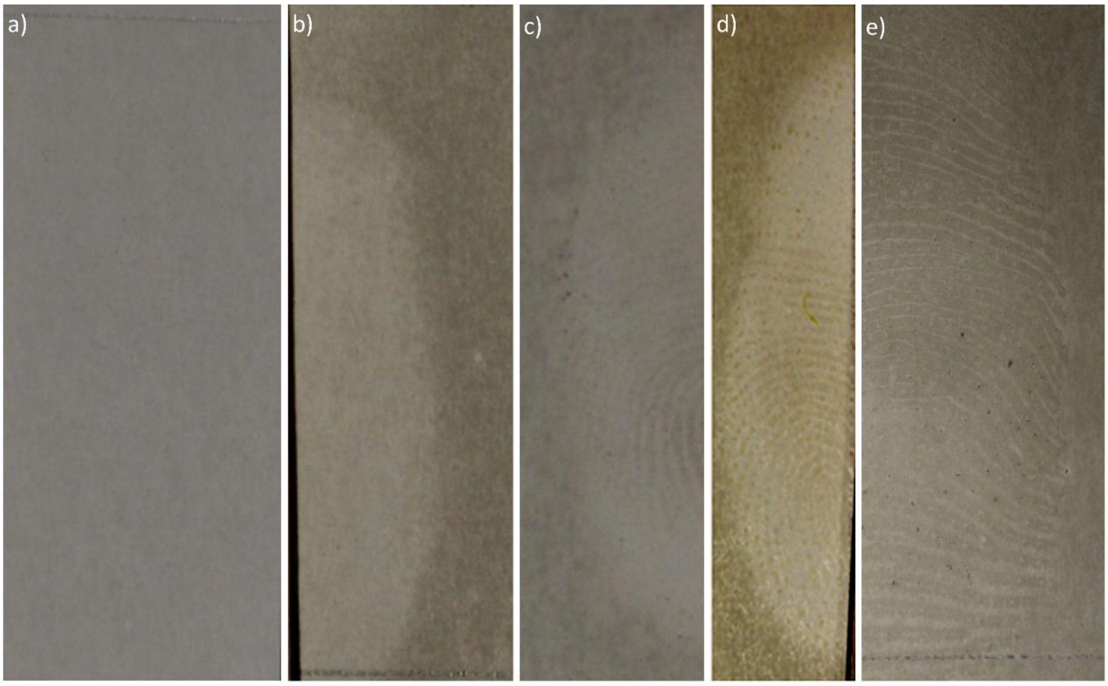

Figure 2a-e: Examples of CAST grades obtained using Au/Zn on thermal paper moving from grade 0 (a) to grade 4 (e). The contrast and brightness has been altered to maximise the detail shown however, no image manipulation has been carried out.

2.2 Study 2: Optimisation of Au/Zn and Ag/Zn

2.2.1 Thermal Paper

To optimise the two different VMD methodologies employed in this study, five different, pristine thermal papers (four white and one with a red spotted pattern) were obtained from a range of retailers. A sheet of each thermal paper was obtained by adhering strips together on the non-thermally active side. Natural fingermarks were deposited on the thermally active side of the thermal paper by six donors for the first paper type (this was increased to twelve donors for the remaining types) in a six mark depletion series using thumbs, index and middle fingers from both hands with a line denoting where each mark was to be split. The donors were split approximately 70:30 females to males, with an age range from 20 to 60. For the first paper type, three replicates were used; however, the method was updated during this study, and this changed to one replicate when more donors were used. This alternative to replicates is commonly employed within the fingermark enhancement research community and is outlined in guidelines by the IFRG, as is the utilisation of a six mark depletion series for porous substrates [36].

2.2.2 Metals and Mark Optimisation

To determine the optimal weight of gold or silver to be used, a range of lengths, from 1.5mm to 7mm, of both metals were cut and the weights of each accurately measured using a Mettler Toledo NewClassic ML analytical balance to ±0.1mg. This was used as a quality control measure where 0.2mm of gold or 0.1mm of silver approximately equates to 0.2mg. By using weights as opposed to lengths, the reproducibility of the techniques can be monitored. The split marks were then developed and graded using Au/Zn and Ag/Zn VMD, as detailed in section 2.1.3, with the exception that the entirety of the silver wire was evaporated in the optimisation and comparison studies whereas the sequential treatment trial followed the FVM and manufacturer's guide, stopping the evaporation when the mark was visualised [5,33]. The zinc deposition was halted when marks were observed to have been developed for all studies.

2.3 Study 3: Comparison of Au/Zn and Ag/Zn

The two VMD methods under investigation were compared across the five different thermal papers used in this study. The method of depositing samples was the same as in study 1 (2.1.1), except that a ten mark depletion from twelve donors and three replicates were used. This satisfies the criteria of a phase two study outlined by the IFRG [36]. To mitigate the impact of uneven deposition pressure applied by each donor upon the results, the side developed with Au/Zn and Ag/Zn was alternated with each replicate. The optimised lengths of metal previously determined for each thermal paper type were used in this study.

3. Results and Discussion

3.1 Study 1: Order of Sequential Treatment

The results displayed in the graphs (Figure 3a – d) are averages obtained for each technique across all donors at the deposition positions 1 – 10. The results for the first 10 depletions from this study demonstrate that the most effective sequence of techniques for this paper, when taken to completion, is 8% Lumicyano™ followed by Au/Zn and Ag/Zn (Figure 3d). This is also highlighted in Figure 4, where it can clearly be seen that 8% Lumicyano™ as a post-treatment is substantially less effective in comparison to Ag/Zn. It is also apparent from Figure 3a and c that Au/Zn is a more effective method to use as a single process when compared to Lumicyano™ particularly further down the depletion series. This is evidenced by an average grade of between ≈1.2 and ≈1.7 for Au/Zn at the tenth depletion compared to between 0 and ≈ 0.2 for Lumicyano™ at the same point.

Generally, the average grade obtained for Lumicyano™ in Figure 3a decreases with the deposition series as anticipated. This is due to a reduction in the amount of residue deposited as material on the fingertip decreases, leaving less available on the surface upon which the cyanoacrylate based Lumicyano™ polymer can form. Additionally, absorption of the organic components may have occurred; however, the inorganic components, such as NaCl, would be available on the surface which absorbed water during the processing with Lumicyano™ when the relative humidity was approximately 80%. Therefore, this may have provided an initiator for polymer growth [29], which would explain why some development with Lumicyano™ was observed. Due to the almost semi-porous nature of thermal paper, the FVM indicates in primary chart 3, that cyanoacrylate treatment is often the most effective method on a semi-porous substrate when followed by VMD or powder [5]. This is generally backed up by the findings from this research as can be seen in Figures 3a – 3c where the results are consistent with this general statement. Figure 3d, however, indicates that Au/Zn is less effective after Lumicyano™, in contradiction to 3b. This is an anomalous observation as the Au/Zn hs been shown to form around the Lumicyano™ polymer and enhance visualisation [37], however, on polymers the effectiveness of CA+VMD has been shown to be substrate dependent [38]. This decrease in grade after Lumicyano-Au/Zn treatment may have been caused by differences in fingermark deposits related to a variable that would affect all donors, such as the weather being significantly different on the day of deposition. However, as the sequences starting with Au/Zn (Figure 3a and 3c) show similar grades to each other, on the different deposition days, the

Lumicyano™-VMD interaction is potentially more inconsistent or more affected by variability in processes. Therefore, the multi-process Lumicyano™-VMD sequence may require tighter process tolerances than the individual visualisation techniques.

When the final treatment of either Ag or Ag/Zn is applied, there is a substantial difference in the results obtained. It can be seen that when Ag is the final process in this sequence, there is a decrease from the average grades observed with Au/Zn, with the exception of the 10^{th} depletion where there was no change (Figure 3b). Whereas, Ag/Zn as a final technique (Figure 3d) resulted in a substantial improvement in the average grade of marks from ≈0.2 to ≈1.3, which is likely due to the increased contrast obtained by using zinc. When silver is then applied after Au/Z (Figure 3a) the average grade of marks remains fairly consistent with those obtained for Au/Zn (with the exception of position 1 to 4 where Ag increased the development observed). This indicates that Ag is more effective for heavier deposits. However, for the sequence shown in Figure 3c, Ag/Zn did not improve the average grade of marks observed with the exception of the first depletion. This could be attributed to poorer contrast being achieved due to overdevelopment of with zinc (as described in the FVM [5]). For both Figure 3a and 3c, it is apparent that Lumicyano™ has a deleterious effect on the average grade of marks when applied as the final process on this paper type. This is likely due to limited residue being available around which Lumicyano™ could form a polymer and as such, should not be used. This is contrary to a study which investigated alternative VMD metals on polymers and proposed a sequence of Au/Zn VMD followed by Ag VMD then cyanoacrylate fuming [39]. However, the results observed were in agreement with the FVM as this indicates that cyanoacrylate fuming is ineffective after VMD [5]. It is clear that Lumicyano™ as a post treatment is ineffective on this paper type and should be avoided.

As was observed from this study, the most effective sequence of techniques was Lumicyano™, Au/Zn, then Ag/Zn (Figure d). However, this is only true when the sequence is taken to completion. Examining the average of the ten mark depletion series with individual single-process techniques, Au/Zn developed marks of average grade 1.60 compared to Lumicyano™ which developed marks with an average grade of 0.47. A similar analysis shows that Au/Zn with Lumicyano™ pre-treatment averaged 0.81. Therefore, it was determined that Au/Zn VMD was the single most effective technique to use. It is hypothesised that the Lumicyano™ as a primary treatment had a detrimental influence on the contrast obtained by the Au/Zn method, however the second treatment of Ag/Zn then overcomes this issue. As a result of these observations, the decision was taken to focus on single treatments in subsequent studies.

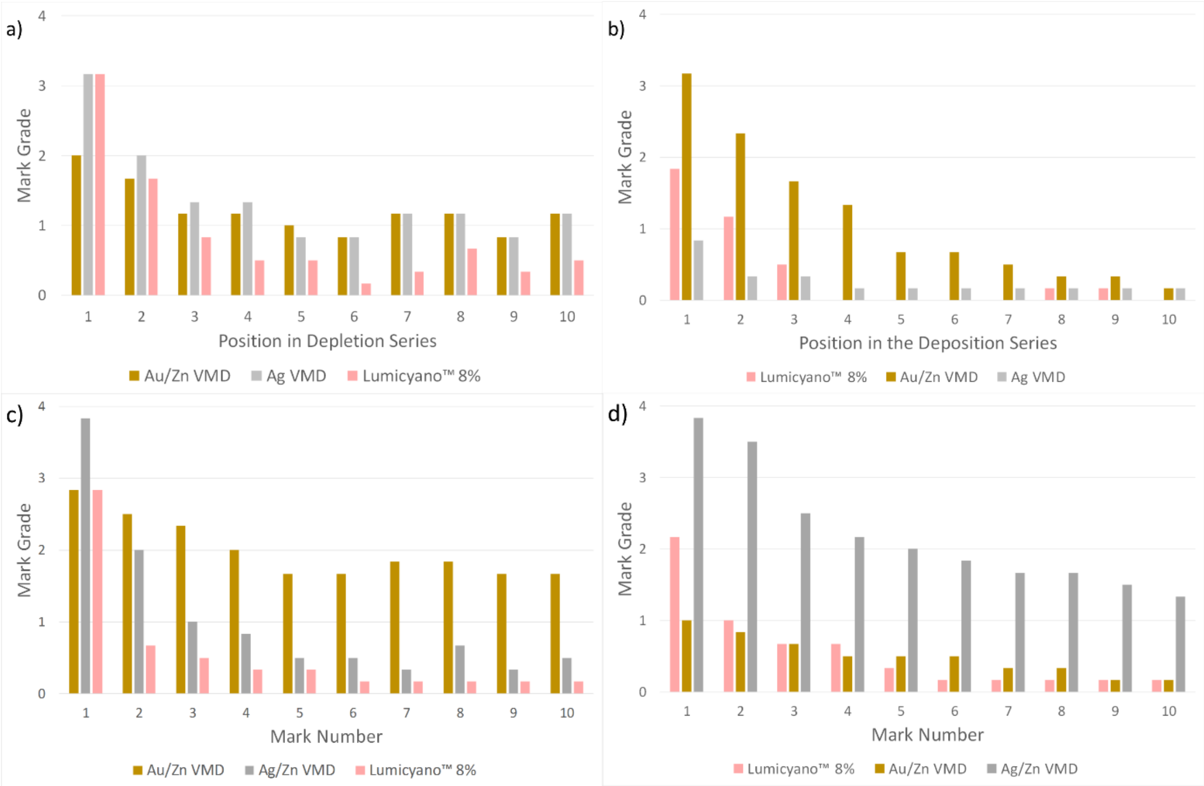

Figure 3a – d: Graphs of the average grade of fingermarks obtained at the first 10 positions after sequential development using the processes under investigation. The marks were graded between each treatment according to the previously described criteria and then the average across the donors was determined. Graphs a and c show the average Au/Zn grade obtained when this process was used as a first technique followed either by (a) Ag/8% Lumicyano™ or (c) Ag/Zn/8% Lumicyano™. Graphs b and d show the sequences with 8% Lumicyano™ as a first technique followed by Au/Zn and either (b) Ag or (d) Ag/Zn.

3.2 Study 2: Optimisation of VMD techniques

The results for the optimisation of Au/Zn and Ag/Zn on different thermal papers (Table 3) shows that the optimum length of gold and silver is variable depending on paper type. The results do not appear to be normally distributed, and the developed contrast is also sometimes poor but dependant on paper type.

Table 3: Summary of the optimal lengths determined for each VMD processes and paper type as determined by identifying the length that gave the greatest percentage of identifiable marks (grades 3 or 4 on the adapted CAST scale).

Paper sample	Au (mm)	Ag (mm)
Paper 1	1.5	3
Paper 2	1.5	3
Paper 3	2	1.5
Paper 4	1.5	4
Paper 5	6	4

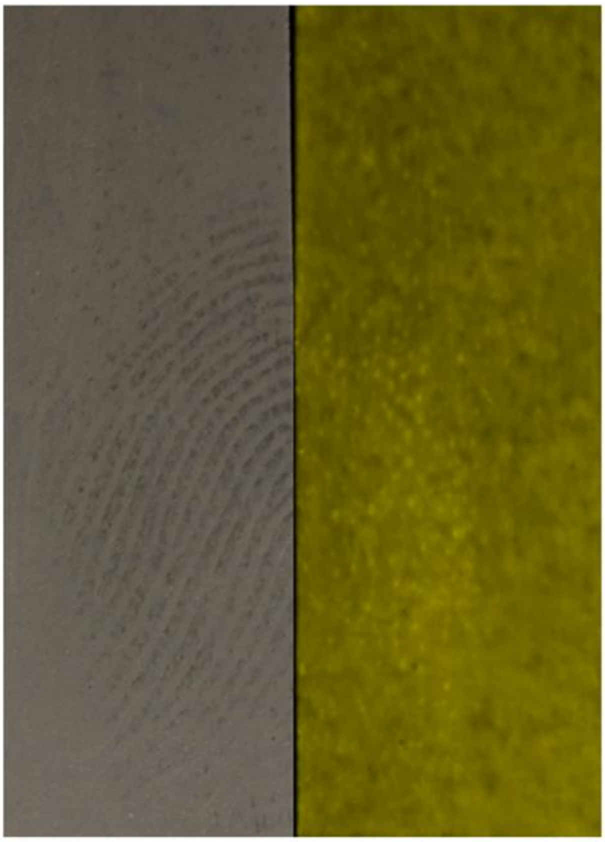

Figure 4: Example of half marks at the first depletion from the same donor at the end of different sequential treatments. Left shows the mark after the Ag/Zn final treatment and the right after an 8% Lumicyano™ final treatment. The differences in terms of ridge detail obtained can clearly be seen.

According to the FVM [5], the most effective length of gold is 3 – 5mm for non-porous substrates however, the optimal length determined experimentally for these five papers falls outside this range. The 6mm of Au that is optimum for paper 5 appears to be separate from the grouping of the other papers at 1.5 – 2mm optimum. This is possibly due to topographical or chemical interactions between the substrate and the fingermark which will be explored further in future work. When compared to the other paper types, paper 3 can be seen to require a different amount of Ag (1.5mm) to achieve optimum development. This could be attributed to the coloured and patterned background of this substrate resulting in better contrast being obtained with less Ag. This study demonstrated that the quality of primary metal development is variable depending upon paper type. For paper 1, for both gold and silver, the difference between the percentage of identifiable fingermarks (grades 3 and 4 on the CAST scale) only varies by approximately 1 – 3% from the optimal length of metal and the surrounding lengths tested. This is in contrast to paper 5 where the deviation from the percentage of identifiable marks was between 0 to 12.5% for gold and 4.2 to 7% for silver demonstrating the variability of results is dependent upon paper type. This research demonstrates the importance of aligning development procedures to the optimum is also substrate dependent.

The observed differences for both metals on each paper type could be attributable to differences in the composition of the surface of the thermal paper. A more hydrophobic coating is likely to result in water soluble components remaining on the surface whereas, a less hydrophobic coating is likely to lead to more of the fingerprint residue being absorbed into the thermal paper which would reduce the effectiveness of VMD as an enhancement technique. Additionally, the topography and surface energy of the different substrates could have an impact on the spreading of the fingermark residue [40] which in turn may influence the amount of gold or silver required to achieve development. It may be that the greater the spread of the residue, the less primary metal is required. Therefore, if a ridge has spread then the residue is covering a greater area but the layer is thinner. This, in combination with absorption into the paper, could mean that larger amounts of the precursor metal result in some agglomerates forming in the ridge which are available to the subsequent zinc treatment. If this is the case, then the contrast between the ridge and the background would be reduced and would explain the results observed.

One aim of this study was to explore using Ag/Zn as a technique in the same way as Au/Zn, where the entire piece of gold is evaporated. The results for Ag/Zn show that this does have potential as a viable technique for fingermarks on thermal paper. This is further explored in the third study which compared standardised methodologies for Ag/Zn VMD enhancement against Au/Zn development, with each process optimised for the individual paper type.

This work also highlighted the potential for very low amounts of gold and silver (1.5mm) to successfully enhance marks. As a result of this, it may be financially beneficial to investigate the potential to utilise a smaller amount of gold or silver on a range of commonly processed surfaces to determine if this yields comparable results.

3.3 Comparison of the two optimised VMD processes

The respective optimised amounts of gold and silver (outlined in Table 3) were compared on the five thermal paper types with the results of this study analysed by determining the representative grade at each position by averaging results from each donor and replicate. From Figure 5 it can be seen that the effectiveness of the VMD processes varies with thermal paper sample, and no one process is clearly most effective across all samples.

From Figure 5a, it is apparent that the Ag/Zn method is more effective at optimising fingermarks on Paper 1, particularly when there is more material deposited on the surface (positions 1 – 3). It is likely that the variation of the Ag/Zn process is due to the decreasing residue down the depletion series whilst the Au/Zn process is less affected by depletion number and the consistency observed is probably due to the amount of gold being a limiting factor. For paper 2 (Figure 5b), Au/Zn is a more effective technique for enhancing marks which is evidenced by Au/Zn developing more marks to an identifiable quality for all but the first depletion. For the first depletion, Ag/Zn proved to be more effective at developing the latent marks (≈28% compared to ≈14%). Whilst for paper 3 (Figure 5c), the two techniques performed similarly with the exception of position 6 where Ag/Zn developed ≈8% compared to Au/Zn developing 0% of marks to an identifiable quality. It is clear that these techniques had limited effectiveness on paper 4 (Figure 5d), the exception being at the first depletion for Ag/Zn where almost half of all fingermarks were developed to an identifiable standard. Whereas on paper 5 (Figure 5e), Ag/Zn is a more effective technique at the first depletion where it results in 50% more marks being enhanced to grade 3 or 4 than the Au/Zn method. Thereafter, Au/Zn is the more effective technique down the rest of the deposition series. As was observed, the Au/Zn technique was very effective on paper 2 as well as paper 5, albeit to a lesser extent, perhaps indicating that the papers are similar in structure and surface characteristics. Scanning Electron Microscope (SEM) images of paper demonstrate both the variable structure of regular paper and variability of the surface texture and composition between different types or brands [41,42]. It can be inferred that variability of surface topography of thermal paper samples in this study affect the fingermark deposition and enhancement [7].

The Ag/Zn process consistently has a greater standard error on the different thermal papers (with the exception of paper 3) as demonstrated by the larger error bars shown on the graphs. These errors highlight the variance observed for the grading of fingermarks and goes some way to explain any apparent irregularity in results, such as increases at positions with lower amounts of residues. Other factors that were not monitored, such as deposition pressure of the mark and donor diet, contributed to both inter- and intra-donor variability. It is also likely that the printed design present from manufacture on paper 3, a pattern which is not uniform over the depletion series, led to increased variability of the contrast obtained which could explain the increased percentage of identifiable marks at positions 4, 7 and 10. It is uncertain whether this printed design was interacting with the fingermark at a chemical level or purely in terms of observable contrast.

For 4 of the 5 paper types tested, Ag/Zn proved to be more effective than the commonly used Au/Zn for the first depletion mark, and the techniques were of comparable efficacy on the fifth paper. As shown in Figure 3, for the first depletion the Ag/Zn process developed approximately 12 – 47% of marks to an identifiable standard compared to approximately 3 – 25% for Au/Zn. Overall across all papers Ag/Zn developed approximately 29% first depletion marks to an identifiable standard, compared to 14% for Au/Zn. This indicates that the silver based VMD technique requires a greater amount of residue to be effective and when more residue is present it is more effective than Au/Zn. However, over the ten depletion series Au/Zn produced 8.3% identifiable marks and Ag/Zn 7.7% across all papers demonstrating that the optimised Ag/Zn technique has comparable effectiveness to the optimised Au/Zn VMD method. Additionally, for papers 1, 3 and 4, the optimised Ag/Zn method had a comparable sensitivity to the optimised Au/Zn process, the selection of technique is therefore strongly substrate dependent.

The average variance between replicates was calculated by determining the variance between the different replicates for each donor and then averaging this value across the twelve donors. Through this method, it can be seen in the supplementary information (Figures S3a – e) that the variance at each position in the deposition for each paper type is typically at or below 0.5 of a grade. There were some exceptions to this, at position 1 with Ag/Zn for each paper type the average variation was consistently between approximately 0.65 and 0.85. Variation of approximately 0.58 was also observed for the Au/Zn method at position one on paper type 3 whilst a variation of around 0.7 was seen at position two with Ag/Zn. Paper two recorded the greatest average variation as Au/Zn was seen to vary by approximately 0.8 to 1 grade for positions two to six. The variation observed may be accounted for by different atmospheric conditions on the day of deposition as well as changes in donor diet and other known intra-donor variability factors [34].

The use of a standardised Ag/Zn method was explored in this study and whilst it was relatively effective (Figures 5a – c), the efficacy of development was substantially dependent upon paper type (Figure 5d and e). The benefits of a standard methodology for Ag/Zn to ensure consistency and reproducibility of development between individuals means that this is an area that would benefit from further research.

Whilst the contrast obtained was sometimes poor, the use of the VMD methods on thermal paper did enhance many marks to an identifiable standard. There remains an issue with photography of the developed marks as the VMD often produced a reflective layer which made visualisation and capture challenging. Future work could investigate the potential of utilising alternative lighting to increase the contrast and reduce the glare, which would make visualisation of the developed marks more effective.

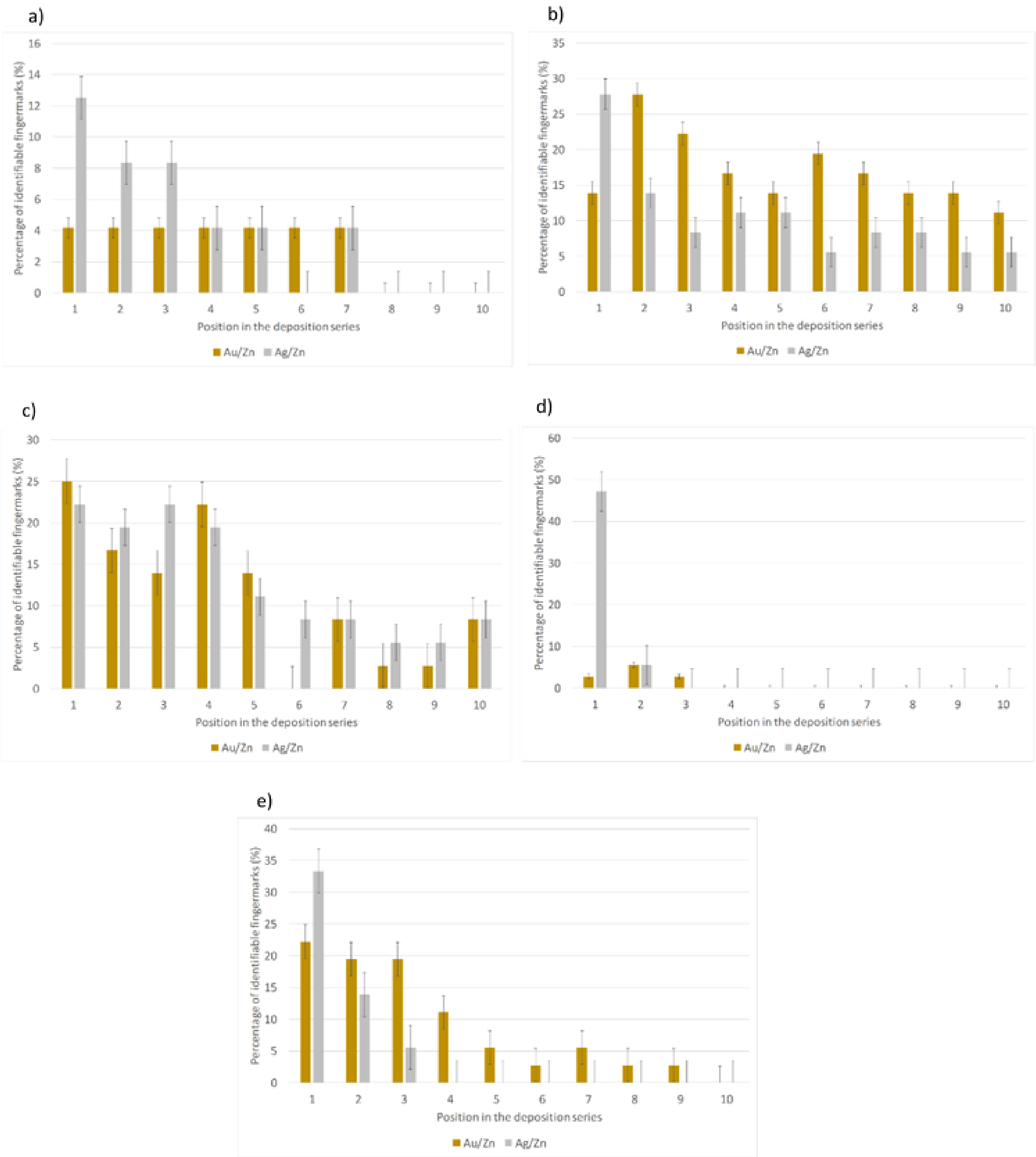

Figure 5a – e: The comparison of optimised Au/Zn and Ag/Zn on (a) paper 1, (b) paper 2, (c) paper 3, (d) paper 4 and (e) paper 5 across a 10 mark depletion series. It should be noted that the scales on each graph are different to allow effective visualisation of the data.

4. Conclusion

When used in series with Au/Zn and Ag, or as a post-treatment after Au/Zn and Ag/Zn, Lumicyano™ is ineffective or deleterious to fingermark enhancement on this type of thermal paper; however, when used as the first technique with Au/Zn and Ag/Zn there is an improvement of the quality of fingermarks but only if the sequence is taken to completion. As a result of this study, Lumicyano™ was deemed to be too variable to be used in sequence with these VMD methods on this substrate.

The two VMD processes, Au/Zn and Ag/Zn, were successfully optimised for five different types of thermal paper however, the amount of gold or silver was found to be dependent upon the substrate. Generally, 1.5-2mm of gold was required except for paper 5 which required 6mm, in contrast to the recommended amount of 2 – 3mm for non-porous surface [5]. Whereas, 3-4mm of silver was the optimum amount for all but one of the paper types, compared to a by-eye visualisation in manufacturer's guidelines. This indicates that other substrates could be investigated with varying amounts of gold and silver, potentially leading to an improved performance and cost savings if smaller lengths of material are more effective than standarad amounts currently used.

Once optimised, the VMD techniques were compared on five different thermal paper types. It was observed that more residue resulted in a greater percentage of marks being enhanced to grade 3 or 4 on the CAST scale when the Ag/Zn method was used, regardless of paper type. For paper types 2 and 5, the Au/Zn optimised method was more effective overall, however, the Ag/Zn optimised method performed better overall on papers 1 and 4 with no noticeable difference between the techniques on paper 3. It is also apparent that both techniques are sensitive to these changes of substrate; and from the results obtained, the choice of technique is dependent upon the substrate. This research also determined that the Ag/Zn technique requires a greater amount of residue present to obtain development than the Au/Zn method. However, when these conditions exist the Ag/Zn process can be the more effective than Au/Zn VMD, in this study developing 29% of first depletion marks to an identifiable standard, compared to 14% for the Au/Zn process.

5. Acknowledgements

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6. Conflict of Interest

There are no conflicts of interest to declare.

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8. Supplemtary information

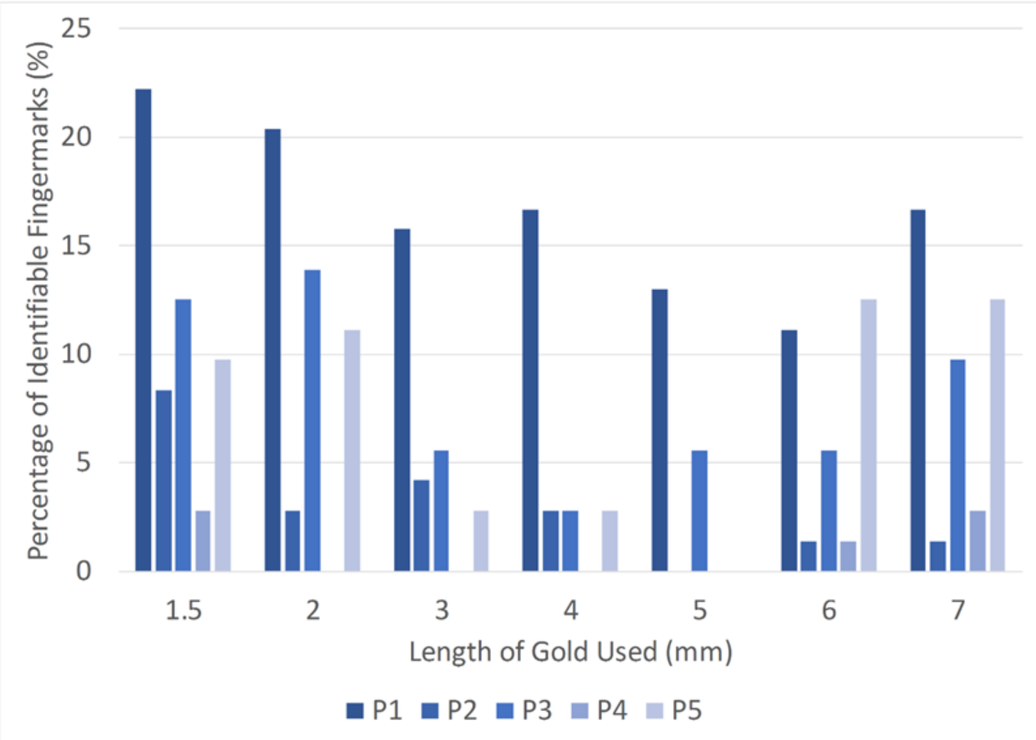

Figure S1: The percentage of identifiable fingermarks (grades 3 or 4) developed on each of the different thermal papers (P1 – P5) using different lengths of gold.

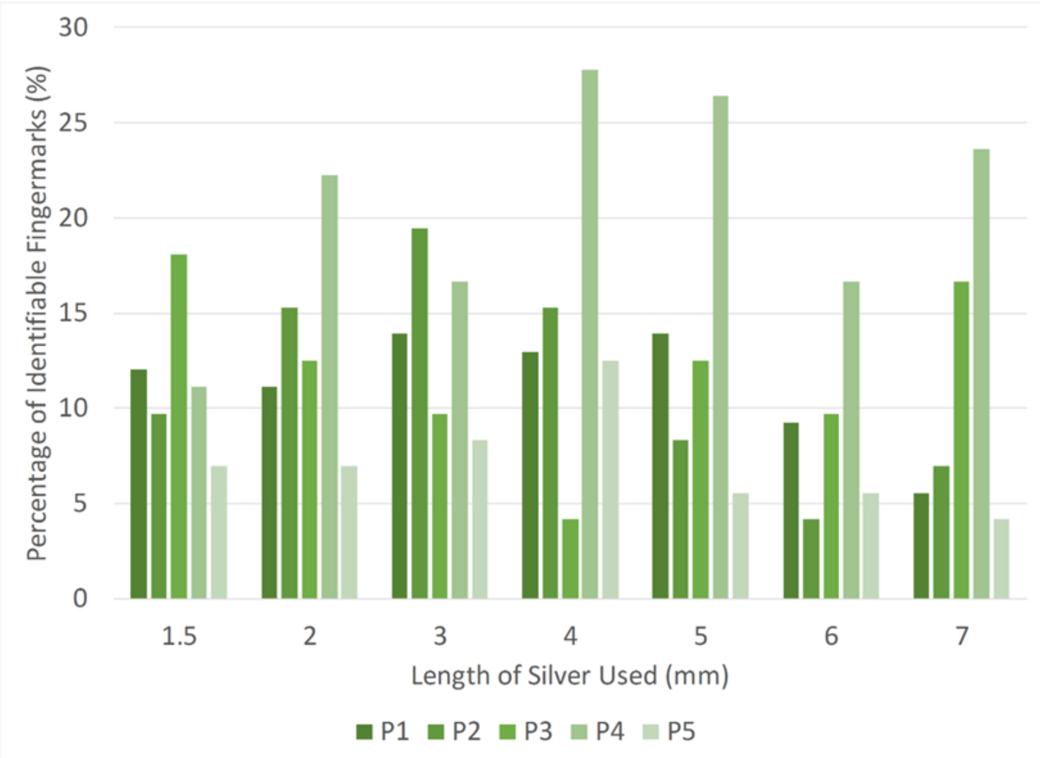

Figure S2: The percentage of identifiable fingermarks (grade 3 or 4) developed on each of the different thermal papers (P1 – P5) using different lengths of silver.

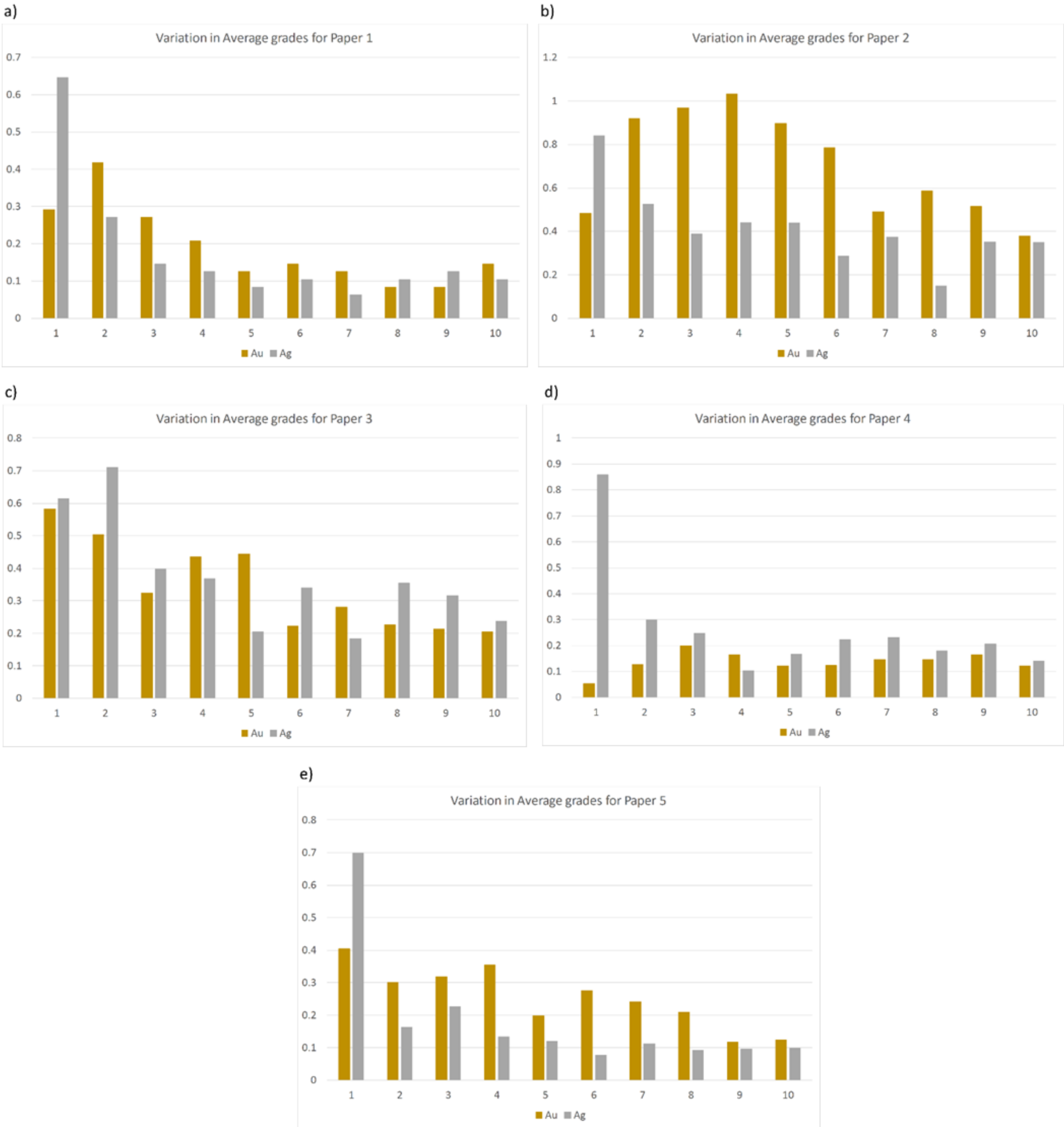

Figure S3a – e: The comparison of variance in grade observed for Au/Zn and Ag/Zn developed marks on all papers across the 10 mark depletion series averaged across donors.